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An investigation on total ozone over western Mediterranean(*)

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Summary. — During recent years ozone depletion has been detected not only over polar regions but also over mid-latitude areas. This study analyzed daily total ozone (TO) data from three south-western European locations in order to detect long-time TO trends by means of a filtering technique. Correlation analysis with atmospheric circulation patterns was carried out to explain the decreasing trends observed. Results appear to show a strong correlation between TO decrease and the North Atlantic Oscillation and Arctic Oscillation Indices throughout recent decades. On the other hand, the trends also indicate that, at least during the last ten years, TO variations cannot be explained solely by natural atmospheric cycles over the studied area.

PACS 92.70.Gt – Climate dynamics.
PACS 92.60.Bh – General circulation.
PACS 95.75.Wx – Time series analysis, time variability.

1. – Introduction

In the last three decades, total ozone has been decreasing in many places, including over Europe. Currently, one of the main subjects of interests is to understand to what extent this decline is due to human activity or rather to natural climatic causes and/or atmospheric circulation variability.

At middle latitudes ozone variations are mainly due to dynamical processes at different time scales which are interpretable in terms of horizontal and vertical transport as well as to the intensity of positive or negative sources. At southern Mediterranean latitudes horizontal stratospheric advection very likely plays a major role [1]. Vice versa, north of the polar jet, stratosphere-troposphere ozone downward flux is active and this shall be accounted for in offering possible interpretations. The authors do not aim at proposing an

(*) The authors of this paper have agreed to not receive the proofs for correction.

overall mechanism able to interpret any ozone change by means of a horizontal advection scheme.

Short-term variations may be associated with weather conditions: in connection with high-pressure systems, the tropopause raises pushing up air out of column with the reduction of ozone [2, 3]. In western Europe an increase of ozone amount is often found in the rear side of cyclones at the passage of a cold front, while a minimum may be found in the south-west side of a high-pressure area close to the warm front [4]. In addition, atmospheric circulation can influence interannual ozone variability and long-term trends. Investigations on ozone trends in terms of the North Atlantic Oscillation (NAO) and the Arctic Oscillation (AO) Indices were carried out by several researchers [5-7]. It has been suggested that a) the predominance of the positive phase of the NAO in the last decades is closely related with this decreasing tendency of ozone and b) the NAO influence on TO features in Europe is expected to be higher than that of the Quasi Biennial Oscillation (QBO) or that of the solar activity.

The paper has an observational and descriptive character confined and concentrated on an area (south-western Mediterranean) not often analysed, without any ambition to propose or suggest new physical mechanisms.

Long-time trends of total ozone (TO) were estimated for three western Mediterranean locations by filtering the long-time ozone series collected at the three stations by means of Dobson solar spectrophotometer.

The relationship between ozone behaviour and circulation indices such as North Atlantic Oscillation index (NAO) and Arctic Oscillation index (AO) was also examined in order to explore any possible linkage between ozone and atmospheric circulation. Trends, as well as correlation, were computed on yearly basis and for winter season when the strongest coupling between troposphere and stratosphere takes place.

2. – Ozone and NAO index trends

2˙1. *Filtering technique*. – According to Rao and Zurbenko methodology, described in Eskridge *et al.* [8] and Milanchus *et al.* [9], any geophysical variable $X(t)$ can be separated into several components as follows:

$$X(t) = e(t) + S(t) + w(t) + T(t) + \varepsilon(t)\,, \tag{1}$$

where $w(t)$ is the meso-synoptic scale component (weather pattern related), $S(t)$ is the seasonal component and $e(t)$ is the long-time scale signal. $T(t)$ and $\varepsilon(t)$ represent turbulence and instrumental error, respectively. These two components were negligible in the case in which the average monthly data are used.

With reference to cycles, $w(t)$ may well be the high-frequency component, $S(t)$ the middle-frequency term (periods from 3 weeks to 1 year), and $e(t)$ is the low-frequency one, with a periodicity of at least one year.

To detect any possible trends, the lowest frequencies have to be isolated. The KZ (Kolmogorov-Zurbenko) filtering technique consists of an iterative moving average effective in removing frequencies higher than a specific threshold. To remove the periodicity shorter than P months, a filter KZ (D, N) was used in order to satisfy the following criterion:

$$D\sqrt{N} \leqslant P, \tag{2}$$

where D indicates the temporal window width and N is the number of iterations.

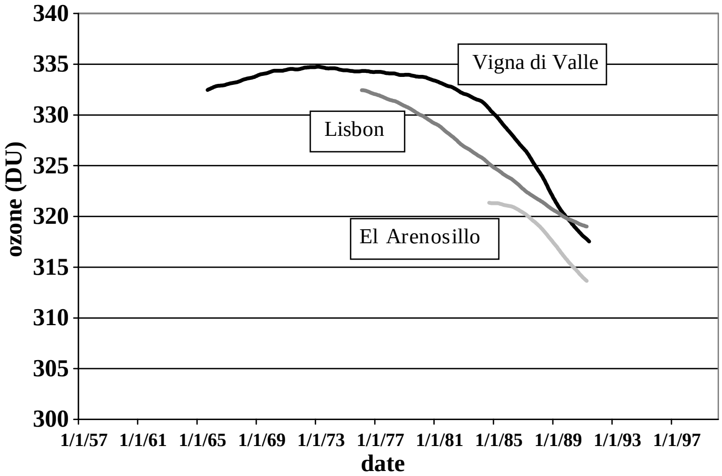

Fig. 1. – Filtered series by using the KZ(105,2).

2˙2. *Total ozone long-term trend.* – All total ozone (TO) data were extracted from the World Ozone and Ultraviolet Radiation Data Center (WOUDC) at Toronto (Canada). The analysed stations were Lisbon, El Arenosillo and Vigna di Valle (table I).

A KZ (105,2) filter was applied to the monthly series in order to remove all periodicity shorter than 12.4 years. In addition to seasonal and annual fluctuations, other known cycles, such as the Quasi Biennial Oscillation (QBO) and the 11-year solar cycle, were eliminated. The resulting filtered series are shown in fig. 1.

It should be noted that use of a different filter, KZ (120,1), saves the 11-year solar cycle, whose contribution to the total ozone series is as follows: 3.4% for Lisbon, 3.0% for El Arenosillo and 2.3% for Vigna di Valle.

Ozone trends were subsequently estimated by using a linear regression analysis on the long-term component series (fig. 1) beginning from the year in which the decline appeared to start (last column in table II). The % trend per decade with the uncertainty is reported in table II. The correlation coefficient R (last column in table II) between the filtered series, $e(t)$, and the remaining series, $\mathrm{TO}(t) - e(t)$, indicates that the specific components are independent.

The decline in total ozone in most European areas started in the 70s; this is also confirmed for Lisbon and Vigna di Valle series, (-2.6 ± 1.2)% per decade and (-2.8 ± 1.1)% per decade, respectively (table II).

For El Arenosillo station the decreasing tendency started only in the early 80s although ozone observations at this station began in the mid 70s and the filter ignored

Table I. – *Stations and period of the series.*

Station	Country	Latitude	Longitude	Height (m)	Period
Lisbon	Portugal	38.77 N	9.15 W	105	Sept. 60-Dec. 99
El Arenosillo	Spain	37.10 N	6.73 W	41	Jan. 76-Dec. 99
Vigna di Valle	Italy	42.08 N	12.22 E	262	July 57-Dec. 99

Table II. – *Total ozone trends with the filter KZ* (105,2).

Station	Estimated decline starting year	Trend %/decade	R
Lisbon	1975	-2.6 ± 1.2	0.041
El Arenosillo	1984	-3.7 ± 1.7	-0.125
Vigna di Valle	1973	-2.8 ± 1.1	-0.003

Correlation coefficient R is given with $p < 0.05$.

data from both ends of the series. Thus data for El Arenosillo cover a shorter period than those of the other two stations. The delay in the decline starting year is presumably due the gap in data series and not to a physical process.

Therefore the trend estimated with the filtered data (KZ (105,2)) may not be accurate and thus the decline might have started earlier.

2˙3. *Ozone winter and NAO index trends.* – Given that circulation patterns change noticeably from winter to summer, a trend analysis limited to the cold period of the year (December to March) was performed. First, the series containing solely winter mean ozone values were retrieved. Subsequently a KZ(12,1) filter was applied on yearly winter values to remove all periods not exceeding 12 years. Results in fig. 2 and table III indicate behaviours quite similar to the monthly data although somewhat less smooth. The trends were obtained by linear regression, starting from the year in which the decline started. The winter trend for Lisbon is slightly weaker than its annual trend (-2.4% per decade for winter, -2.6% per decade for the annually), while the other two stations showed that winter trends were stronger than the yearly trends (table III). The values obtained are in agreement with those reported by WMO [3].

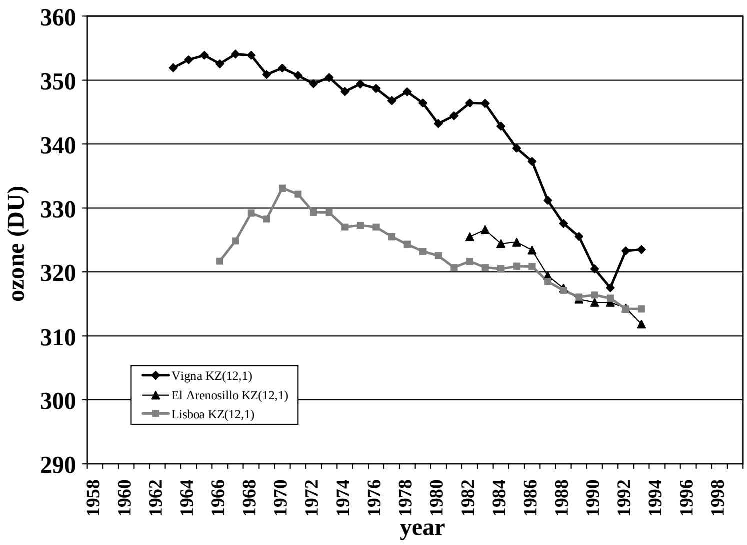

Fig. 2. – Winter series by using the KZ(12,1). Diamond marker line for Vigna di Valle series, grey square marker line for the Lisbon one and triangle marker line for that of El Arenosillo station.

TABLE III. – *Total ozone winter trends with the filter KZ* (105,2).

Station	Time period	Winter trend % per decade
Lisbon	1969-1993	-2.4 ± 1.1
El Arenosillo	1982-1993	-4.6 ± 2.1
Vigna di Valle	1963-1993	-3.5 ± 1.9

Ozone depletion, in addition to the generally accepted concept of being the consequence of the increase of chlorine concentration in the atmosphere, may be related to changes in the general atmospheric circulation specially in the stratosphere as suggested by a recent study [10]. Indeed large variations in the strength of the stratospheric circulation are followed by anomalous tropospheric regimes affecting NAO and AO.

The North Atlantic Oscillation (NAO) is a major disturbance of the atmosphere over the North Atlantic Ocean. It affects to a considerable extent the climate of the near continents from Eastern North America to Western Europe [5]. The NAO is commonly described by an index whose time behaviour gives information on temporal variations of surface pressure difference between a station in the Azores and a station in Iceland. Thus it is related to the changes in the strength of the dominant middle-latitude westerly wind flow during winter. NAO's characteristic is that it alternates yearly between positive and negative index values. Depending on the phase of NAO, the atmospheric pressure distribution changes, as does the tropopause pressure value. In high-pressure conditions, the tropopause raises producing a shrinking of vertical column and hence low ozone levels. Conversely, low tropopause pressures are associated with horizontal convergence, air column stretching and high total ozone column.

During the positive phase, the depression over Iceland is below its normal average, and the Azores pressure is higher than average reinforcing the westerly flow across the Atlantic towards Europe advecting ozone poor maritime air. Vice versa, during the negative phase of the NAO, the westerly flow is weak or even replaced by currents with southward components and hence ozone rich air affects Europe. This determines an anticorrelation between NAO and ozone.

Monthly averaged data from the National Center for Atmospheric Research (NCEP/ NCAR) were used to derive mean NAO Index winter values from December to March.

The NAO index time series was filtered by KZ (12,1) operator, similar to that applied to the TO winter series. Figure 3 shows an increasing NAO trend beginning in 1970. No trend is detectable from 1958 to 1969; the mean value is -0.34, whereas a definite positive tendency, with a mean value of $+0.26$, appears between 1970 and 2000, even if in the graph only the period from 1958 to 1989 is considered.

Comparison of the behaviours of TO and NAO indices (figs. 2 and 3) clearly indicates the anticorrelation of the two series and that the respective trends began around 1970.

3. – Space differences in the ozone behaviour

In order to investigate on space difference in TO behaviour, a correlation analysis with NAO index was carried out with significance level $p < 0.05$.

Correlation coefficients on a monthly basis are shown in table IV. Major importance is attributed to the coefficients in bold with explained variance $> 15\%$ (the explained variance is the percentage of the total ozone variance accounted for by a linear regression

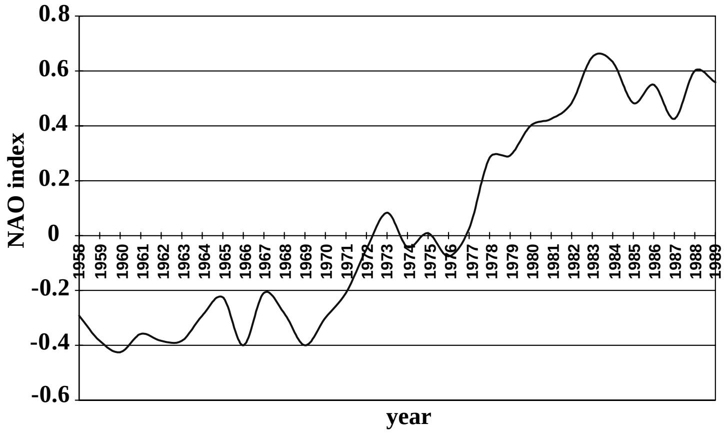

Fig. 3. – NAO Index time series.

model where NAO and AO are independent variables). Although the use of NAO Index is recommended for winter only, coefficients were computed for every month. As expected, ozone and NAO index are anticorrelated. The stronger anticorrelation occurs in February at the Iberian stations, while at Vigna di Valle the maximum is found in March even if at the limit of significance. It is interesting to note that, for the Iberian stations, anticorrelation is also present in April and in January at the El Arenosillo station. At Vigna di Valle the anticorrrelation is very poor in all months.

The differences between Lisbon and Vigna di Valle are remarkable. A possible interpretation is that the western Mediterranean area is more intensively affected by cold outbreaks of ozone-rich air from northern latitudes with respect to the Atlantic coast. This phenomenon is more marked during low NAO index periods in winter.

According to Thompson and Wallace [11] and Baldwin [12], the Arctic Oscillation (AO) is the circulation annular mode that carries information on the intensity of the arctic

Table IV. – *Correlation coefficients between TO mean values and those of NAO and AO indices.*

	Lisbon		El Arenosillo		Vigna di Valle	
	NAO	AO	NAO	AO	NAO	AO
January	−0.277	−0.299	**−0.462**	**−0.388**	−0.382	**−0.570**
February	**−0.452**	**−0.412**	**−0.506**	**−0.390**	−0.323	−0.291
March	−0.317	*	−0.260	−0.365	**−0.383**	**−0.514**
April	**−0.413**	*	**−0.409**	*	*	*
May	−0.288	*	−0.318	*	*	*
June	−0.210	*	*	*	*	*
July	*	*	*	*	*	*
August	−0.230	*	*	−0.249	*	*
September	−0.338	−0.294	*	*	*	*
October	−0.317	−0.201	*	*	*	−0.273
November	−0.221	*	−0.235	*	−0.203	*
December	*	*	−0.266	0.382	*	*

polar vortex. AO index data are available for the period 1958-2000 (NCEP/NCAR). The monthly correlation between NAO and AO indices from 1959 to 2000 is high mainly in winter ($R = 0.780$).

As in the case of NAO index the correlation between TO and the AO index series was computed (table IV). Winter correlations are homogenous as far as sign is concerned at the three stations. The fact that the Mediterranean area is more prone to cold outbreaks from higher latitudes than the Atlantic Iberian coast, presumably leads to a greater AO index at Vigna di Valle.

4. – Concluding remarks

The aim of the present study is to investigate long-time trends of total ozone (TO) and their relationship to atmospheric circulation in the western Mediterranean region. So far the total ozone in this area was not particularly studied in relation to circulation indices. The type of filter analysis used was effective in detecting trends in the long time series of total ozone and NAO index.

Results show the existence of a remarkable ozone downward tendency over the western Mediterranean, with a decrease of about 3.0% per decade. The result agrees with recent studies [3]. The cause of this trend is not completely understood so far. Continuity equation for atmospheric ozone indicates that total ozone time changes may be due to three-dimensional transport and intensity of sources. Although the latter is often mentioned in the literature as the main cause of ozone depletion (chemical consequences of chlorine increase) the effect of circulation changes, particularly within the stratosphere, requires further studies.

Moreover, it is still unclear whether NAO positive trend is simply the result of natural variability or an effect of the global warming.

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